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**APPLICATIONS OF TUNABLE HIGH ENERGY/PRESSURE PULSED LASERS  
TO ATMOSPHERIC TRANSMISSION AND REMOTE SENSING**

**By**

**R. V. Hess and R. K. Seals, Jr.**

**September 1974**



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# APPLICATIONS OF TUNABLE HIGH ENERGY/PRESSURE PULSED LASERS TO ATMOSPHERIC TRANSMISSION AND REMOTE SENSING

By R. V. Hess and R. K. Seals, Jr.  
Langley Research Center

## SUMMARY

Atmospheric transmission of high energy  $C^{12}O_2^{16}$  lasers can be considerably improved by pulsed high pressure operation which, due to pressure broadening of laser lines, permits tuning the laser "off" atmospheric  $C^{12}O_2^{16}$  absorption lines. Pronounced improvement is shown for horizontal transmission at altitudes above several kilometers and for vertical transmission through the entire atmosphere. The atmospheric transmission of tuned  $C^{12}O_2^{16}$  lasers compares favorably with  $C^{12}O_2^{18}$  isotope lasers and CO lasers. The advantages of tunable high energy, high pressure pulsed lasers over tunable diode lasers and waveguide lasers in combining high energies with a large tuning range are evaluated for certain applications to remote sensing of atmospheric constituents and pollutants. Pulsed operation considerably increases the signal/noise ratio without seriously affecting the high spectral resolution of signal detection obtained with laser heterodyning.

## INTRODUCTION

Recently new types of tunable lasers have been developed with wide tuning ranges ( $> 3$  GHz) for pulsed operation at high pressures ( $> 1$  atmosphere) and high energies ( $> 10$  Joules). These tunable high energy/pressure lasers (e.g., review in Ref. 1) have received considerable attention because of their potential application to laser produced fusion, optical radar, isotope separation, and commercial uses. Their applications to atmospheric laser energy transmission and remote sensing of constituents and pollutants, however, have not previously been evaluated although the wide tuning range at high laser energies offers considerable promise. Such an evaluation, as attempted in this paper, must, of course, be partly based on comparison with the application of other laser sources. The paper thus presents new theoretical studies for specific examples coupled with a synthesis of information from diverse fields such as high energy lasers, tunable lasers, atmospheric spectroscopy, laser energy transmission, and remote atmospheric sensing.

Efforts are in progress at NASA and other laboratories to improve the atmospheric transmission of high energy gas lasers operating with  $CO_2$ , CO, DF, and HF by extending their wavelength range into regions of reduced absorption by atmospheric constituents such as  $H_2O$ ,  $CO_2$ ,  $O_3$ ,  $CH_4$ ,  $N_2O$ , and others. In this paper quantitative theoretical evidence is given that considerable improvement in atmospheric laser transmission can be attained with high

energy/pressure pulsed lasers since these lasers can be tuned across the pressure broadened laser lines which can be made wider than atmospheric constituent absorption lines. This improvement is especially significant for horizontal transmission at altitudes above several kilometers and for vertical transmission through the entire atmosphere. Calculations are performed for transmission of tunable high pressure CO<sub>2</sub> lasers operating at low energies. Nonlinear effects are not considered; however, the improved transmission from the linearized calculations indicates the potential importance of tuning these lasers at high energies. In view of linear reversibility the calculations apply to "up" and "down" transmission between various altitudes (e.g., sea-level, mountains, aircraft) and orbiting satellites, the shuttle, or other spacecraft. Improvement in transmission for tunable high pressure pulsed CO lasers is briefly discussed also.

Applications of low power tunable lasers to remote sensing of atmospheric constituents and pollutants from the ground, aircraft, satellite, or shuttle are being investigated by several groups. In References 2 and 3 applications of CW tunable diode lasers are summarized; these lasers have the very wide tuning range, narrow line widths, frequency stability, and fine tuning capability desired for remote sensing, but their CW power is in general presently limited to several mW although higher powers of 0.25 W have been obtained in some cases. Reference 4 discusses the application of waveguide lasers (Refs. 5,6,7) which attain high gain and high pressures through use of narrow diameter laser discharge tubes. CO<sub>2</sub> waveguide lasers have been operated at powers approaching 2 W, but presently the pressures for CW operation are limited to  $\approx 0.5$  atmospheres, corresponding to a line width of  $\approx 1.5$  GHz and a tuning range (off line center) of  $\leq 0.75$  GHz. This is still below the requirements for several important remote sensing experiments discussed in this paper. Pulsed waveguide lasers have been operated in isolated cases at pressures approaching one atmosphere (Ref. 8), with a correspondingly increased tuning range.

High energy/pressure tunable pulsed lasers which are not restricted to narrow discharge tubes, are capable of operation with both the wide tuning range and the high energy required for many important transmission and remote sensing experiments. The tuning rate of high pressure lasers across pressure broadened laser lines is of the order of 3 GHz/atmosphere, e.g., for CO<sub>2</sub> lasers with typical high pressure gas mixtures. As pressures of 10 atmospheres are approached, overlapping of the laser lines occurs and the tuning becomes continuous. In order to attain sufficient frequency stability for fine tuning and narrow line widths, the production of uniform plasmas is of great importance and preionization by UV photons (Refs. 9,10,11) or electron beams (Refs. 12,13) is desirable. The photo-preionization techniques discussed in References 9 and 10 promise to be especially effective for combining plasma uniformity with compactness of the laser device.

Acknowledgement is made of beneficial discussions with F. Allario and P. Brockman.

## SYMBOLS

c	speed of light
E	energy
h	altitude
$\hbar$	Planck's constant
k	absorption coefficient
P	laser power
S	signal at detector
T	post detection integration time
$\beta$	heterodyne detection bandwidth
$\nu$	frequency
$\bar{\nu}$	wave number
$\tau$	transmittance

### Subscripts:

o	CW operation
p	pulsed operation

### Abbreviations:

CW	continuous wave
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## ATMOSPHERIC TRANSMISSION OF TUNABLE HIGH ENERGY/PRESSURE PULSED $C^{12}O_2^{16}$ AND $C^{12}O^{16}$ LASERS

All of the calculations presented here have been performed using a line-by-line absorption and transmission computer program described elsewhere (Ref. 14). In addition, continuum absorption (Ref. 15) by  $H_2O$  and  $N_2$  has been included. A mid-latitude summer model atmosphere has been assumed. The comparative effects of changes in laser frequency and laser altitude on horizontal atmospheric transmittance are conveniently presented in terms of the absorption coefficient  $k$  in  $km^{-1}$ . Since  $k$  is related to the transmittance  $\tau$  through

$$\tau = e^{-\int_0^L k d\ell}$$

it is representative of horizontal transmittance, without having to specify the length  $L$  over which the transmittance occurs. Vertical transmittance is obtained by integrating over the absorption coefficients at various altitudes.

The importance of tunable  $C^{12}O_2^{16}$  lasers for increasing atmospheric laser transmission is shown in Figure 1 for the P(20) laser line centered at  $10.5911 \mu m$ . As indicated by the reduction in calculated absorption coefficient, the advantage in tuning off the line center increases with altitude due to reduction of pressure broadening with increasing altitude. Figure 2 presents the dependence of calculated absorption coefficient vs altitude for the P(20)  $C^{12}O_2^{16}$  laser at line center and tuned 5 GHz off line center (corresponding to a wavelength change from  $10.5911 \mu m$  to  $10.5930 \mu m$ ). The calculated absorption coefficient for the  $10.5248 \mu m$  wavelength of the  $C^{12}O_2^{18}$  isotope laser is also presented for comparison. The considerable reduction in absorption coefficient achievable by tuning the  $C^{12}O_2^{16}$  laser gives absorption coefficients comparable to those of  $C^{12}O_2^{18}$  isotope lasers. However, it may not be desirable to use a  $C^{12}O_2^{18}$  laser due to the expense of the  $C^{12}O_2^{18}$  isotope.

In Figure 3 the altitude dependent reduction in calculated absorption coefficient is shown for the tuned  $C^{12}O_2^{16}$  laser operating at  $10.5930 \mu m$ , a  $C^{12}O^{16}$  laser at  $4.8638 \mu m$ , an HF laser at  $2.9643 \mu m$ , and a DF laser at  $3.8206 \mu m$ . The tuned  $C^{12}O_2^{16}$  laser transmission compares well with each, with the exception of the chemical DF laser. However, the DF chemical laser is presently not capable of closed cycle operation, important to several missions. It must be also noted that the advantages of the DF laser for high altitude transmission above 5 km are not great. Since the  $C^{12}O_2^{16}$  laser is a potential competitor of the  $C^{12}O_2^{16}$  laser for operation in an electric discharge mode with closed cycle operation, it should be mentioned that the  $C^{12}O^{16}$  laser wavelength at  $4.8638 \mu m$  in Figure 3 corresponds to the 2-1 P(15) transition which has a comparatively small atmospheric absorption or high transmittance. Such low vibrational transitions generally operate at lower energies (e.g., Ref. 16), and tuning of some higher transitions in high energy/pressure  $C^{12}O_2^{16}$  lasers may be advantageous for improved atmospheric transmission at high energies.

For practical purposes it is of special interest to plot the vertical transmittance of the tunable high energy/pressure  $C^{12}O_2^{16}$  laser from an altitude  $h$  through the entire atmosphere vs frequency difference  $\nu - \nu_0$  from line center. Figure 4 indicates that the transmittance from  $h = 0$  (ground and  $h = 2.5$  km or 6.5 km (mountains or aircraft) can be considerably increased by tuning 5 GHz off the line center. However, from altitudes  $h = 2.5$  and 6.5 km even half this tuning range yields vertical transmittance which should be useful for certain applications of laser energy transmission. Naturally, for the low energy linearized calculations illustrated here, the results apply equally to transmittance from the top of the atmosphere to altitude  $h$ , which is of interest for applications with the laser situated on the shuttle or satellite. It should be noted that applications of high energy/pressure tunable lasers concern not only transmission of high pulsed energy but also have the potential of transmitting high average powers through use of high repetition rates, based on high flow rates (e.g., Ref. 1).

In order to tune 5 GHz "off" line center a  $C^{12}O_2^{16}$  laser line width of at least 10 GHz is required since the centers of the laser line and the absorption line coincide. Since the line broadening per atmosphere laser pressure is  $\approx 3$  GHz, a laser pressure of  $\approx 3$  atmospheres would be desirable; for horizontal and vertical transmission from higher altitudes, however, even half the tuning range at half the laser pressure would be sufficient. The question also arises as to how much loss in power results from tuning off line center. Since the laser line width is here defined as "full width at half intensity" of an absorption or emission line, tuning over the full width represents a reduction of absorption or emission to half the value. The decrease in laser power with frequency off line center, however, need not be as large since the laser power is influenced by the rate of energy fed into a region of the pressure broadened line, which can be controlled by proper cavity design. The laser power for a given wavelength off line center may be further increased by increasing the pressure of the lasing medium since both line width and laser power increase with pressure (Ref. 1, IV B). Further experiments are required to determine gain and power variations across pressure broadened laser lines.

## SOME APPLICATIONS OF HIGH ENERGY/PRESSURE PULSED TUNABLE LASERS TO REMOTE ATMOSPHERIC SENSING

### Statement of Problems

In Reference 2 a variety of applications of tunable lasers to remote atmospheric sensing are studied. An especially strong relation exists between the area of transmission of tunable  $C^{12}O_2^{16}$  lasers and a remote sensing technique discussed in References 2, 4, and 14 and references therein. This technique is remote sensing by laser differential absorption using diffuse reflection from the earth. The term differential implies a reference laser line "off" the constituent or pollutant absorption line as well as one "on"

the absorption line; the difference in atmospheric transmittance between the "on" and "off" lines is a measure of the pollutant. In view of its strong relation to the preceding transmission problems of tunable high energy/pressure  $C^{12}O_2^{16}$  lasers, the differential absorption technique is illustrated for remote measurement of  $C^{12}O_2^{16}$  concentrations, which is one of the cases discussed in Reference 14. The calculations in Reference 14, however, apply to low pressure  $C^{12}O_2^{16}$  lasers for which the tuning range due to line broadening is very small and different  $CO_2$  laser lines are required to obtain measurements "on" and "off" wavelengths, respectively. The  $C^{12}O_2^{16}$  R(8) laser line at  $9.3418 \mu m$  and the  $C^{12}O_2^{18}$  P(18) isotope laser line at  $9.3414 \mu m$  were chosen for the "on" and "off" wavelengths, respectively. The tunable high energy/pressure  $C^{12}O_2^{16}$  laser has the important advantage of being able to scan the  $C^{12}O_2^{16}$  line, thereby obtaining a dependence of its pressure broadening with altitude and further provides high energy pulses, which as subsequently shown, improve the signal/noise ratio over CW operation. The dependence of pressure broadening on altitude can be used in an inversion technique to obtain a vertical profile of the  $C^{12}O_2^{16}$  concentration in the atmosphere (or more general constituent or pollutant concentrations).

The laser power requirements, of course, depend on the altitude; thus, for example (Ref. 14), for use from the shuttle ( $\approx 200$  km) 25 watts of CW power would be required just to obtain the average or total burden concentration of  $CO_2$ . If in addition, vertical spatial resolution due to pressure broadening of  $CO_2$  is to be obtained, the need to distinguish between  $CO_2$  concentrations at various altitudes brings with it a requirement for higher power. In Reference 14 calculations were performed for a variety of constituents and pollutants such as  $CO_2$ ,  $O_3$ ,  $C_2H_4$ , NO, and CO, whereby a "two-step" profile was obtained for CO and NO from aircraft altitudes of 5 km. The profiles were calculated assuming the use of a low power CW tunable diode laser which has a very wide tuning range (Refs. 2,3). The calculations are extended to high energy/pressure pulsed tunable lasers, later in this paper.

#### Required Tuning Range

The requirement for laser tuning range in laser differential absorption measurements varies for each pollutant, depending on the proximity of laser lines to "on" and "off" positions for atmospheric absorption lines. Some of these problems are discussed in References 2,3,4 and references given therein. Our studies indicate that absorption lines of several constituents or pollutants (e.g.,  $O_3$ ,  $NH_3$ ,  $C_2H_4$ ,  $SO_2$ ) are within 1.5 GHz tuning range from line centers of  $C^{12}O_2^{16}$  and  $C^{12}O_2^{18}$  lasers. It appears that some lines of certain gases (e.g.,  $O_3$ ) require only half this tuning range, but many pollutants

require tuning of at least 1 GHz, making a tuning range of 1.5 GHz from line centers very desirable. This in turn requires line widths of 3 GHz corresponding to a laser pressure of  $\geq 1$  atmosphere. It is presently difficult to attain operation at these pressures in waveguide lasers but such pressures are easily achievable in pulsed high energy/pressure lasers. The tuning range of 1.5 GHz is easily achievable for tunable diode lasers, but presently their power limitations restrict them to certain applications. Tunable diode laser spectroscopy is, however, vital for determining the required tuning range for various pollutants.

The tuning range required of CO lasers ("off" line center) for remote sensing of NO is  $\approx 1$  GHz according to recent tunable diode laser experiments by K. Nill, M.I.T. Lincoln Laboratory (personal communication). This tuning range, corresponding line width ( $\approx 2$  GHz), and laser pressure ( $\approx 2/3$  atmosphere) are, at present, marginally obtainable even for CO<sub>2</sub> waveguide lasers, which are further developed than CO waveguide lasers. These conditions are easily attained in tunable diode lasers. Direct sensing of CO would be possible if high energy/pressure pulsed CO lasers could be operated on the  $1 \rightarrow 0$  transition (Ref. 16). However, in view of high potential efficiencies at high powers, frequency doubling of tunable high energy/pressure C<sup>12</sup>O<sub>2</sub><sup>16</sup> and C<sup>12</sup>O<sub>2</sub><sup>18</sup> lasers offers important possible alternatives for CO- and other pollutant-lines in this wavelength range.

#### High Pulse Power or Energy with Heterodyne Detection

The importance of higher power for increasing the signal-to-noise ratio of the (diffusely) reflected return signal is given in Reference 14 for the case in which the return signal is heterodyned with a local laser oscillator. For a "two wavelength case" with signals "on" and "off" the absorption line the ratio of differential signal  $\Delta S$  to noise  $N$  given in Equation (18) of Reference 14, can be expressed in the form

$$\frac{\Delta S}{N} \propto P \left( \frac{T}{\beta} \right)^{1/2}$$

It is indicated that  $\Delta S/N$  increases with  $P$  and  $(T)^{1/2}$  or with  $E/(T)^{1/2}$  where the energy  $E = PT$ . The attainment of large  $\Delta S/N$  without excessive integration times has considerable advantages for aircraft, satellite, or the shuttle when, due to high speeds relative to the ground, atmospheric or ground conditions may change for long integration times. Here, the pulse power is estimated for the limiting case in which the total integration time equals the duration of a single pulse by setting  $(\Delta S/N)_p$  equal to  $(\Delta S/N)_o$ ; for the more general case when one sums over a number  $n$  of pulses the time  $T$  becomes  $T_{\text{sum}} = T_p n$ . There follows

$$P_o(T/\beta)_o^{1/2} = P_p(T/\beta)_p^{1/2} \quad \text{or} \quad P_p = P_o(T_o/T_p)^{1/2} (\beta_o/\beta_p)^{1/2}$$

Using, for example, the CW values  $P_o = 25 \text{ W}$ ,  $\beta_o = 10^6 \text{ Hz}$ ,  $T_o = 10^{-1} \text{ sec}$  for calculations (Ref. 14) of total  $\text{CO}_2$  column content from  $\approx 200 \text{ km}$  altitude (shuttle) and assuming the values  $\beta_p = 10^{-6}$  and  $T_p = 10^{-5}$  for pulsed operation, there results for  $P_p$

$$P_p = 2500 \text{ W}$$

For comparison of pulsed and continuous operation an average power for a repetitively pulsed system is calculated. Assuming a pulse duration  $T_p$  of  $10^{-5} \text{ sec}$  and a repetition rate of 100 pulses per second for 2500 W pulse power, the pulse energy is  $2.5 \times 10^{-2} \text{ joule}$  and the average power 2.5 W. The repetition rate of 100 p.p.s. is assumed since it should give at least comparable atmospheric coverage (from fast moving aircraft or space vehicles) to that obtained for a  $10^{-1} \text{ sec}$  integration time with CW operation. Pulsed high energy/pressure lasers are thus easily capable of attaining the same  $\Delta S/N$  and atmospheric coverage at considerably lower average powers than continuous lasers.

It must be emphasized that although high energy/pressure pulsed tunable lasers offer great potential for long range remote sensing, the present and potential usefulness of CW diode lasers and CW and pulsed waveguide lasers is of great importance to remote sensing. CW operation of 1 mW single mode is no longer exceptional for diode lasers and considerably higher powers should be available in the future over a wide wavelength range covering many absorption lines. Furthermore, while 1 mW single mode CW operation may be marginal for differential absorption measurements from high altitudes using diffuse reflection from the earth, it is sufficient for several applications using long path differential absorption in the atmosphere and heterodyning of laser and passive radiation with the tunable diode laser as local oscillator (Refs. 2 and 3). Operation of pulsed waveguide lasers at pressures approaching 1 atmosphere has been demonstrated (Ref. 8), and further studies are being performed for CW and pulsed waveguide lasers to obtain stable operation at higher pressures, with a correspondingly increased tuning range. The future optimum configurations for given remote sensing missions may actually consist of various combinations of tunable diode, waveguide, and high energy/pressure pulsed lasers. In this manner, small high energy/pressure tunable lasers, with special flashlamp configurations for preionization, may be developed or stable low power diode- or waveguide-lasers may be used as master oscillator matched to a high energy/pressure laser used as power amplifier in an M.O.P.A. laser system.

Finally, since on several occasions concern has been expressed that use of pulses may seriously reduce the benefits of heterodyne detection, this problem is briefly discussed. The building of a wave packet with a short time interval  $\Delta t$  requires superposition of waves with a large spread in frequencies  $\Delta \nu$  (e.g., Ref. 17). This is expressed in the form  $\Delta \nu \geq 1/\Delta t$  and is also the basis of Heisenberg's uncertainty principle  $\Delta E \Delta t \geq \hbar$  with  $E = \hbar \nu$ . In the present case  $\Delta \nu$  and  $\Delta t$  are identified with  $\beta$  and  $T$  thus, for  $\beta = 10^6$  Hz,  $T \geq 10^{-6}$  sec. Since  $\beta$ , the heterodyne detection bandwidth, is considerably smaller than the width of constituent or absorption lines, even at high altitudes, pulsed operation does not seriously affect the high spectral resolution of heterodyne detection.

### CONCLUDING REMARKS

Atmospheric transmission of high energy  $C^{12}O_2^{16}$  lasers can be considerably improved by pulsed high pressure operation which, due to pressure broadening of laser lines, permits tuning the laser "off" atmospheric  $C^{12}O_2^{16}$  absorption lines. The improvement is pronounced for horizontal transmission at altitudes above several kilometers and for vertical "up" or "down" transmission through the entire atmosphere. A large increase in transmittance is obtained by tuning the laser 5 GHz "off" line center which requires a laser line width  $\geq 10$  GHz corresponding to laser pressures  $> 3$  atmospheres. However, even at half the tuning range (corresponding to half the pressure) the vertical transmittance appears sufficient for vertical transmission from and to high altitudes. The transmittance of the tuned  $C^{12}O_2^{16}$  lasers is comparable to or better than that of  $C^{12}O_2^{18}$  isotope lasers and  $C^{12}O^{16}$  lasers operating at low vibrational transitions. Tunable high energy/pressure pulsed lasers have important applications to remote atmospheric sensing of constituents and pollutants and for several applications offer significant advantages over tunable diode lasers and the CW waveguide lasers. Pulsed operation considerably increases the signal/noise ratio for laser remote sensing without seriously affecting the high spectral resolution of signal detection obtained with laser heterodyning.

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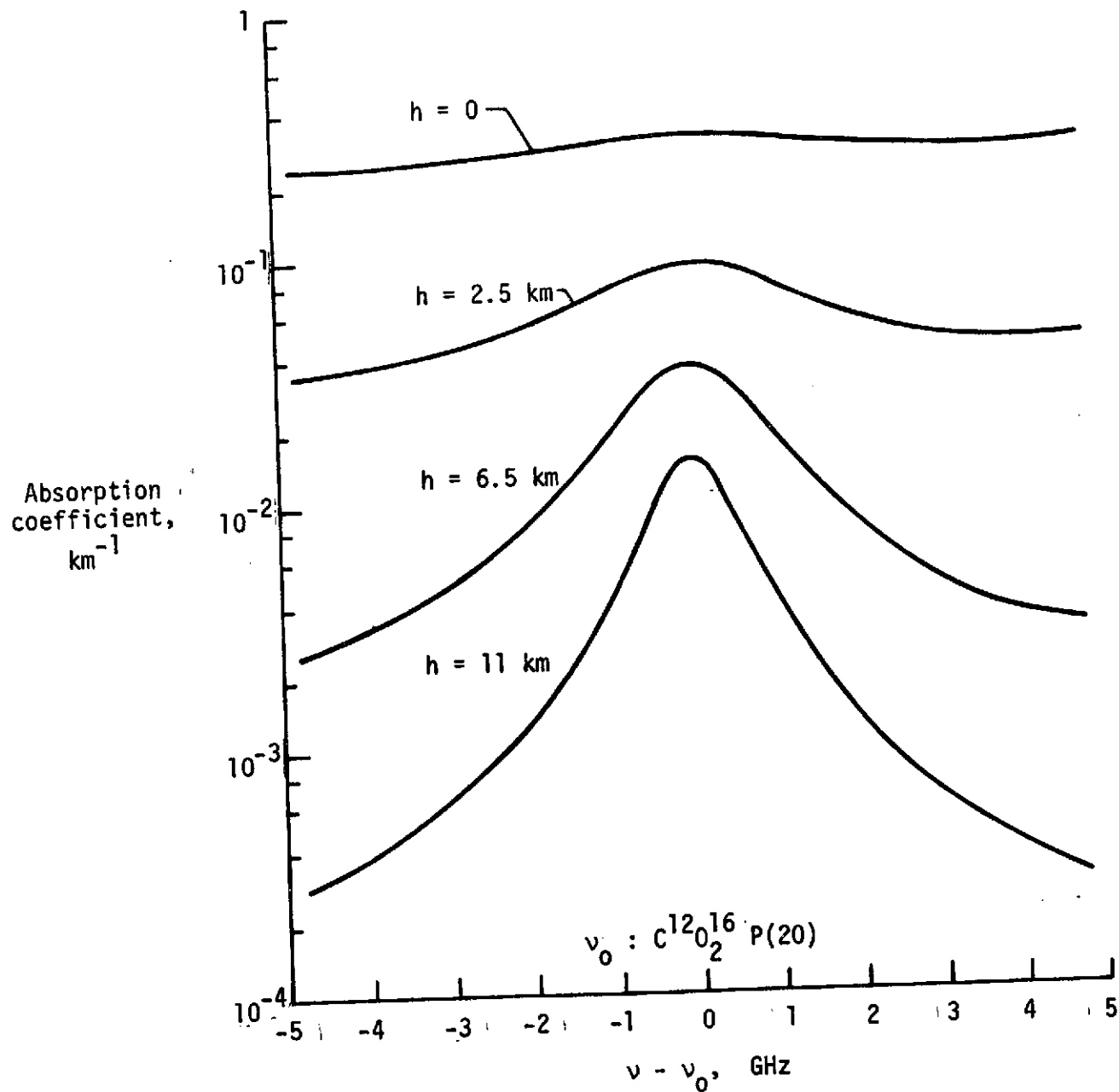


Figure 1.- Calculated frequency variation of absorption coefficient with altitude.

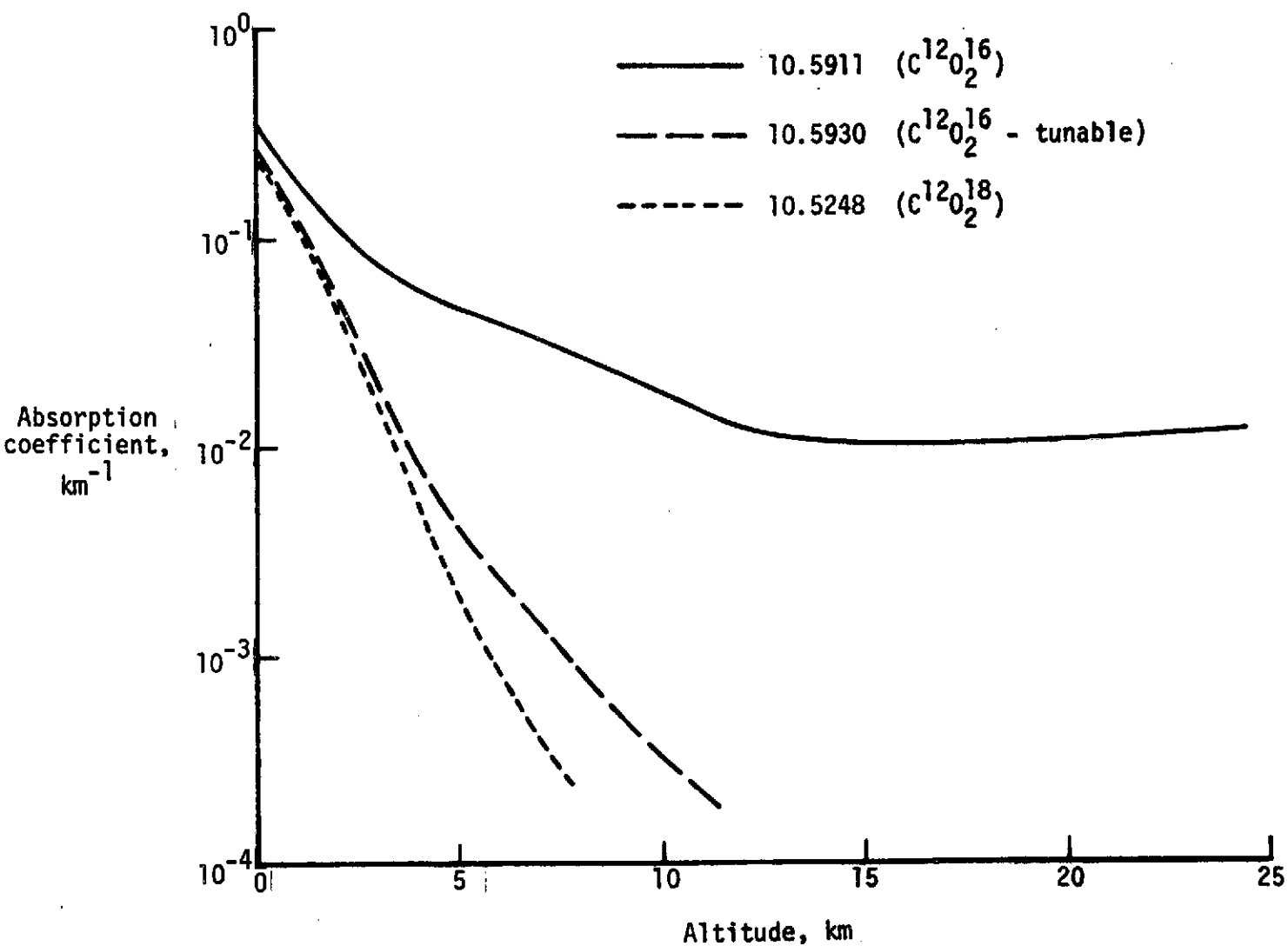


Figure 2.- Comparison of altitude dependence of absorption coefficients for  $\text{CO}_2$  laser types.

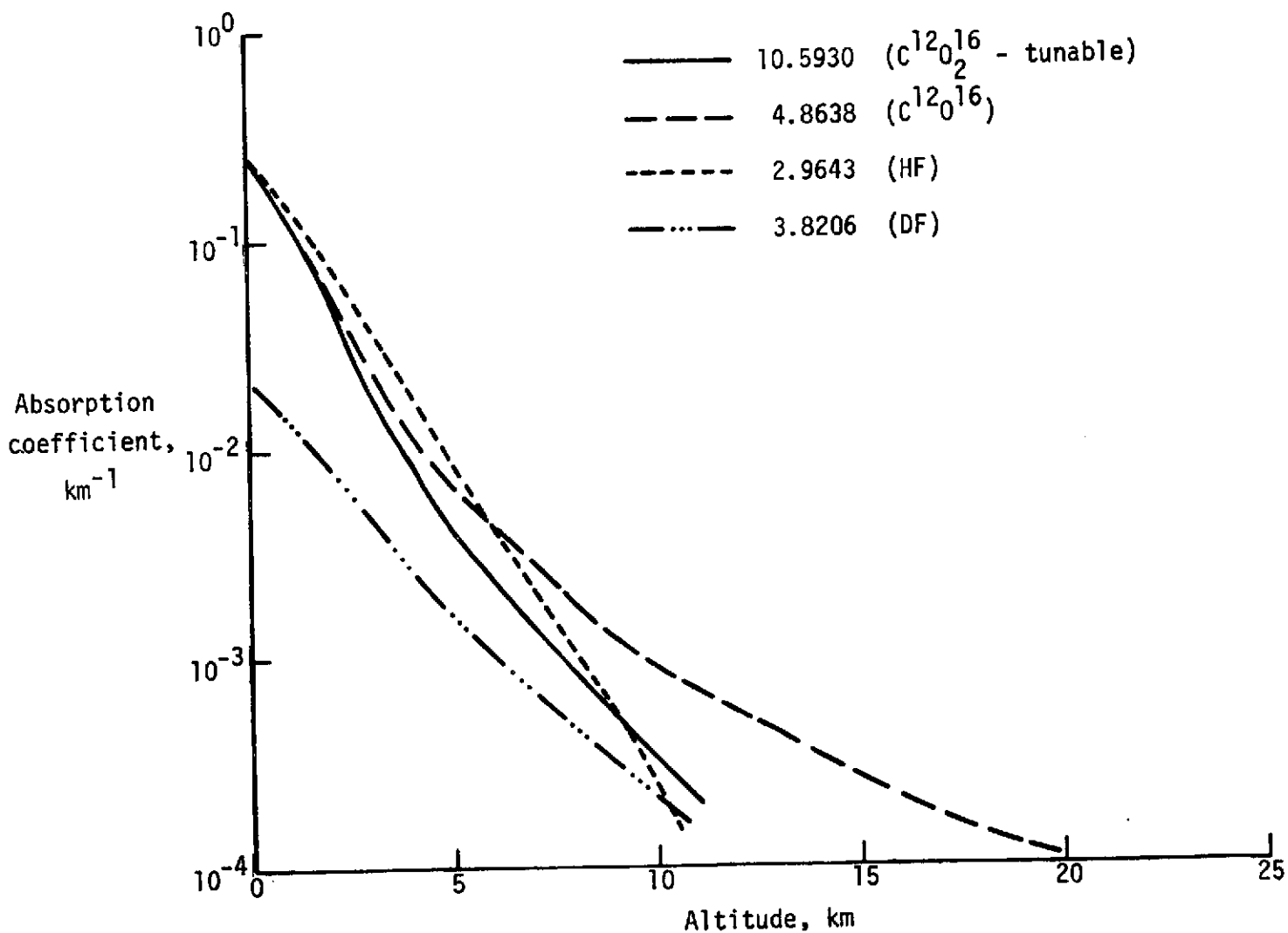


Figure 3.- Comparison of altitude dependence of absorption coefficients for tunable  $\text{C}^{12}\text{O}_2^{16}$  laser with CO, HF, and DF lasers.

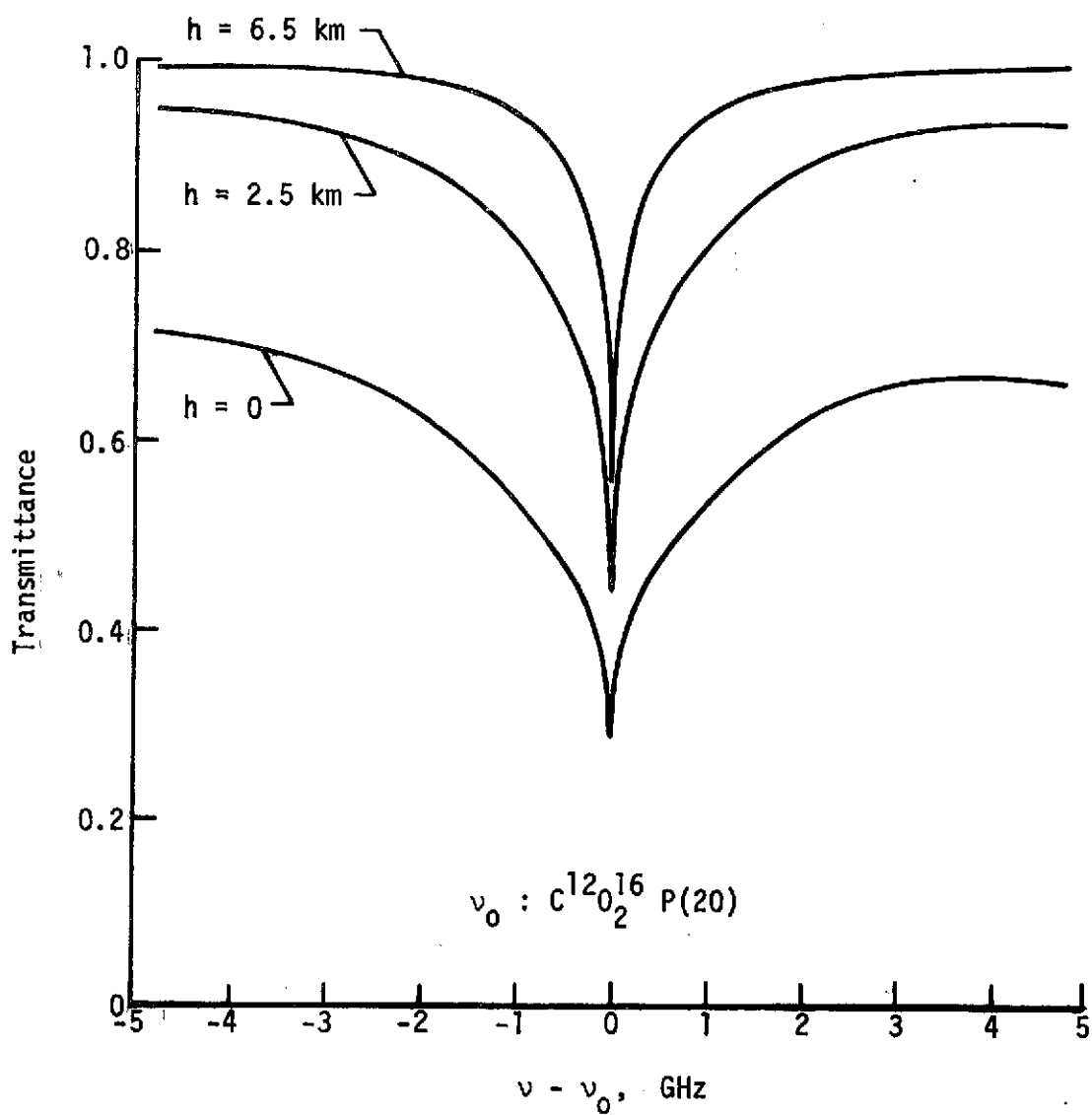


Figure 4.- Calculated frequency variation for tunable  $C^{12}O_2^{16}$  laser for vertical transmittance from various altitudes.